Characteristics of Supercritical Transitional Temporal Mixing Layers

Nora Okong'o and Josette Bellan

Jet Propulsion Laboratory, California Institute of Technology

Pasadena, CA 91109

June 20, 2001

Abstract

Several Direct Numerical Simulation realizations of supercritical, three-dimensional, temporal mixing layers are used to investigate aspects of interest to turbulent combustion. The realizations are based on a model previously developed that accounts for the enlarged transport matrix at supercritical conditions (Soret and Dufour effects), for real gas equations of state, and for variable Schmidt and Prandtl numbers. This model is exercised for two very different sets of binary species, heptane/nitrogen and oxygen/hydrogen, to obtain transitional states for mixing layers excited at different perturbation wavelengths. Visualizations of the transitional states show that all layers develop convoluted regions of high density gradient magnitude (HDGM), which result both from the distortion of the initial density stratification boundary and from mixing. The species mass fraction in these HDGM regions is very weakly dependent of the perturbation wavelength. The existence of the HDGM regions, independent of the perturbation wavelength, indicates that they may be a feature of spatial mixing layers, and furthermore have a similar mass fraction composition. Evaluations of the applicability of the assumed PDF method for describing supercritical transitional flows shows that neither the β density nor the Gaussian are appropriate representations of the conserved scalar, and that the PDFs of the partial densities and the temperature are correlated, invalidating the typical model of the reaction rate as the product of the marginal PDFs.

Key words: supercritical transitional mixing layer

1 Introduction

The turbulent mixing of fluids at high pressure is a topic of much interest because it is relevant to many combustion applications. Such applications include general automotive (particularly, Diesel), gas turbine and liquid rockets engines. Liquid rocket combustion conditions present a particular experimental challenge as the operating regime is supercritical with respect to both fuel and oxidizer and the mixture is potentially explosive. In this situation, numerical simulations with validated models can contribute information that would be otherwise impossible to obtain experimentally.

The modeling of supercritical fluid behavior has been so far primarily addressed in the context of drop modeling, bypassing the turbulent aspect of the flows of practical interest. Among such numerous studies, the investigation of Harstad and Bellan [3] is noteworthy because the model was validated using the data of Nomura et al. [7]. The model in [3] is based on Keizer's fluctuation-dissipation (FD) theory [4] which is totally consistent with non-equilibrium thermodynamics. This set of equations was mathematically closed with real gas equations of state, and high-pressure transport properties.

Fundamental simulations devoted to supercritical shear flows have not been attempted until very recently (see [6], [8] and [10]). Such fundamental simulations are clearly necessary to unravel the specific aspects of supercritical turbulent shear flows. A specific aspect recently observed by Chehroudi et al. [1], [2], and by Mayer and Tamura [5], is the lack of jet atomization under supercritical conditions; instead of drop formation, finger-like structures protrude from the jet.

The present investigation is conducted through Direct Numerical Simulation (DNS) using the validated set of conservation equations of [3], described in Section 2, used here in the context of a three-dimensional temporal mixing layer, as in [6], [8], that is perturbed for the purpose of achieving transition. Aspects of the transitional states pertinent to supercritical turbulent mixing and combustion are emphasized in Section 3. Finally, in Section 4, we summarize our findings and state conclusions.

2 Governing equations

2.1 Conservation equations

The differential conservation equations have been discussed in detail by Harstad and Bellan [3] and by Miller et al. [6] for a binary species system, and consist of the continuity, momentum, total energy and species equations. Their important characteristic is the expression of the energy flux and species mass fluxes according to the FD theory, whereby additional to the typical Fourier and Fick's terms encountered at low pressure, the transport matrix is enlarged to contain the Dufour and Soret terms, respectively. The Dufour term is the sum of two contributions each proportional either to the mass fraction gradient, or the pressure gradient, whereas the Soret term contains two contributions each proportional either to the temperature gradient, or the pressure gradient. The addition of the Dufour and Soret terms introduces a new transport process, the thermal diffusion with its corresponding transport property, the thermal diffusion factor, that must be known in order to solve the equations.

To close the system of equations, we select a modified form of the Peng-Robinson equation of state (EOS) [11], [6], as this EOS portrays in a simple manner the behavior of real gases. All thermodynamic properties are consistently calculated from the EOS. One such important property is the mass diffusion factor, α_D, which in this formulation multiplies the mass diffusion coefficient, D, in the Fick's law. The significance of α_D is to indicate departures from mixture ideality ($\alpha_D = 1$), meaning that the molecular mixing is impeded.

2

2.2 Transport properties

In real gases, transport properties are strong functions of the thermodynamic variables and it is thus important to accurately portray the values of $Sc = \mu/\rho\alpha_D D$, the Schmidt number, and of $Pr = \mu C_p/m\lambda$, the Prandtl number, so as to correctly render mixing and heat transfer (here μ is the viscosity, ρ is the density, C_p is the molar heat capacity at constant pressure, λ is the thermal conductivity and m is the mixture molar weight). To this end, Sc and Pr have been accurately calculated using thermodynamic variable dependent properties, and, for computational efficiency [6], at fixed pressure, p, they have been correlated as functions of temperature, T, and mass fractions ($Y_1, Y_2 = 1 - Y_1$). All simulations presented herein have been performed with thermodynamic variable dependent Sc and Pr. On the other hand, for typical fluids μ is too small to enable resolution of all scales in a domain large enough to keep the Knudsen number in the continuum regime, and thus to ensure validity of the Navier-Stokes equations. To palliate this problem, the strategy is to enlarge μ but keep realistic values of velocities, densities and the Reynolds number, Re, so as to depict the correct dynamics while solving the problem in a domain large enough to be in the continuum regime. Finally, depending on the species set, the thermal diffusion factor is found either by comparing simulations with data (see [3] for heptane/nitrogen) or by considering similarity in molecular characteristics (see [12] for oxygen/hydrogen).

2.3 Configuration, and boundary conditions

The configuration is that of a three-dimensional (3D) mixing layer, depicted in Fig. 1 for one of the sets of studied species. The lower stream contains the higher density fluid (subscript 2), and the initial conditions are supercritical (i.e. $p > p_c$ or/and $T > T_c$, where the subscript c denotes the thermodynamic critical point) for the species in both streams. The boundary conditions are periodic in the streamwise, x_1, and spanwise, x_3, directions, and of outflow type for a real gas (boundary conditions derived by Okong'o et al. [9]) in the cross-stream direction, x_2.

3 Results

The studied species and their properties are listed in Table 1. The initial conditions are listed in Table 2. All simulations are conducted with an initial convective Mach number, $M_{c,0} = 0.4$. The simulations are started with streamwise and spanwise vorticity perturbations (whose amplitude is proportional to F_{3D} and F_{2D}, respectively) superimposed on the mean initial velocity profile that is smoothed near the density discontinuity boundary using an error function; see [6] for details. The initial vorticity thickness, $\delta_{\omega,0}$ ($\delta_\omega(t) = \Delta U_0/(\partial U/\partial x_2)_{max}$ where U is the mean (or $x_1 - x_3$ planar average) flow in the streamwise direction, and $\Delta U_0 = U_1 - U_2$ is the velocity difference across the layer), is 6.859×10^{-3} m for all simulations. The only difference between realizations for the same species set is the value of the perturbation wavelength, λ_1, and the size of the domain that is chosen to accommodate four wavelengths in the streamwise and spanwise directions and two vortex pairings. For each set of species, the value of $Re_0 = (0.5(\rho_1 + \rho_2)\Delta U_0\delta_{\omega,0})/\mu_R$ (μ_R is a reference viscosity) for the second simulation is chosen by matching $Re_0\lambda_1/\delta_{\omega,0}$ of the first simulation. This

leads to similar values of $\mathrm{Re}_m = \mathrm{Re}_0 \delta_m / \delta_{\omega,0}$ (where δ_m is the momentum thickness) at transition, which are listed in Table 2. The transitional state is chosen to correspond to the peak in the timewise variation of the volume averaged positive spanwise vorticity. Since the initial mean velocity profile is such that all initial spanwise vorticity is negative, the appearance of positive spanwise vorticity is indicative of small scale formation. Braid and between-the-braid visualizations of the spanwise vorticity concur with the choice based on this global quantity.

To ensure that all DNS herein are well resolved, the energy spectra, both streamwise and spanwise, were computed at the transitional time in a manner similar to Kim et al. [13]. A sample of the results is displayed in Fig. 2 for C_7H_{16}/N_2, and it clear that there is a ratio of more than ten orders of magnitude between the low and high wavenumbers, thereby showing that most of the energy resides in the large scales and that there is no accumulation of energy in the small scales; for O_2/H_2 this ratio is even larger.

The examination of the transitional states focus on the role of molecular diffusion during mixing and on the utilization of the assumed PDF method for the description of supercritical mixing layers.

3.1 The role of molecular diffusion

Molecular diffusion is a very important process even in fully turbulent reacting flows because it is the mechanism that brings molecules of fuel and oxidizer in contact, thereby allowing chemical reactions. A typical measurement of molecular mixing [14] is the product thickness, $\delta_p = \int \int \int_V \rho Y_p dV$, based on the evolution of a conserved scalar, $Y_p = 2\min(Y_1, Y_2)$. This is because the conserved scalar may be interpreted as the product of an infinitely fast reaction, thus indicating the efficiency with which the reactants mix at the molecular scale, i.e. diffuse. On Fig. 3, δ_p is illustrated versus $t^* = t\Delta U_0 / \delta_{\omega,0}$ for all four realizations listed in Table 2. Diffusional mixing is initiated as soon as the layer starts to roll up and it is enhanced following each pairing, as shown by the slope of δ_p. For the C_7H_{16}/N_2 layers, the global molecular mixing seems more effective at the same t^* than for O_2/H_2, this being attributed to the smaller initial ρ_2/ρ_1 that allows earlier pairing; however, this global molecular mixing is not necessarily indicative of local molecular mixing, as will be shown below, and it is precisely the local process that determines the reaction efficiency. For a given set of species, the realization featuring the higher molecular mixing is that having the larger λ_1 (the situation is not immediately clear for O_2/H_2), this being attributed to the longer time necessary to pair during which diffusion has increased effectiveness.

In previous studies of C_7H_{16}/N_2 [8] and O_2/H_2 [12] layers excited at a single wavelength it has been shown that a salient characteristic of supercritical mixing layers is the formation of regions of high $|\nabla\rho|$ (HDGM) which resulted both from the convolution of the initial density stratification boundary and from mixing. These regions were shown to contain partially mixed fluids and to act as material interfaces and damp the formation of small scales, thereby impeding the achievement of a transitional state. To determine the state of molecular mixing at transition, braid and between-the-braid visualizations of $|\nabla\rho|$ were examined as in [8] and [12] to elucidate their composition and state of mixture ideality. Results from a similar analysis are here displayed in Fig. 4a for the braid plane, and epitomize the situation for the between-the-braid plane as well (not shown). The plots illustrate conditional averages of the mass fraction of the heavy fluid (Y_2) with respect to a cut-off

4

value of $| \nabla \rho |$ which is a fraction of the maximum value of $| \nabla \rho |$ in that plane, $| \nabla \rho |_{\max}$; as this cut-off value increases, one probes deeper into the HDGM regions. Noteworthy, for a specified set of species $| \nabla \rho |_{\max}$ increases with decreasing λ_1, this being attributed to the smaller domain of the simulation which produces flows with higher vorticity. Since the stirring frequency is proportional to the vorticity, a higher vorticity induces more stirring while diffusional processes are unaffected by this change. An enhanced stirring at otherwise unchanged molecular diffusion is responsible for the larger $| \nabla \rho |_{\max}$. The results portrayed in Fig. 4a show that the fluid in the HDGM regions is mostly composed of the higher density species, with small amounts of the lighter fluid dissolved into the heavier one, and that the deeper one probes into the HDGM regions, the higher is Y_2. For a specified set of species, the average Y_2 is quite insensitive to the value of λ_1, indicating that these features (i.e. HDGM regions and their mass fraction composition) might persist for spatial mixing layers. Moreover, the value of the average Y_2 seems also insensitive to the set of species, indicating that it might be a generic aspect of supercritical mixing layers at a specified p_r. Comparisons between this average Y_2 and its stoichiometric value (0.89 for O_2/H_2 and 0.067 for heptane/air) reveals that the composition of the HDGM regions is very close to stoichiometric in the O_2/H_2 layer, but very far from it in the C_7H_{16}/N_2 layer (the addition of O_2 is believed to change only slightly this aspect), indicating that O_2/H_2 is much better suited for combustion than heptane/air. This conclusion is confirmed by the results plotted in Fig. 4b where conditional averages of α_D are plotted, similarly to the plot in Fig. 4a. Clearly, as one probes deeper into the HDGM region of the C_7H_{16}/N_2 layer, the mixture becomes more non-ideal ($\alpha_D < 1$) meaning that it has difficulty mixing at the molecular level, whereas the HDGM regions of the O_2/H_2 layer contain fluid close to ideality (well mixed regions at the molecular level). Thus, despite the global indications from δ_p that the C_7H_{16}/N_2 layer mixes better than the O_2/H_2 one, the local analysis reveals that it is precisely the opposite, in that it is the slowest characteristic time of the local diffusional processes that governs the fate of the molecular mixing.

3.2 Assumed PDF modeling

When using assumed PDF models, there are two issues of concern: (1) the form of the PDF, and (2) the value of the moments necessary to construct the PDF. Here we are concerned only with the first issue, and will assume that exact values of the moments are available ('best' situation) by constructing the PDF with DNS extracted moments. Since the β density has typically been used for describing the evolution of the conserved scalar from the segregated state of the species to complete mixing while the Gaussian (whose first two moments are 0 and 3) has been used to describe the fully mixed state, the present study focuses on these two PDFs.

As a typical example, in Fig. 5 we display for one of the four realizations the DNS extracted PDF, the Gaussian and the β density calculated with the DNS extracted moments; plots from the other realizations exhibit similar features. The PDFs are calculated at several cross-stream locations of the layer so as to illustrate the mixing features of the layer. Clearly, neither the β density nor the Gaussian are appropriate representations of the conserved scalar due, in particular, to the peaks in the DNS extracted PDF. These peaks are a consequence of the HDGM regions characteristic of these supercritical mixing layers.

Because the examined databases were created without including chemical reactions, they can be used on a 'best' case basis to reveal information about the modeling of the reaction rate, which is a joint temperature - (partial densities) PDF, by the product of the marginal PDFs (a common assumption in assumed PDF reacting flows). Indeed, when including reactions, the PDFs will be additionally correlated, further invalidating the replacement of the joint PDF by the product of the marginal PDFs. Among the four realizations, we illustrate in Fig. 6 the comparison between joint and product of the marginal PDFs which appeared the most favorable. Even in this situation, it is apparent that the comparison does not validate this frequent modeling assumption. The discrepancy between the joint PDF and the product of the marginal ones is attributed to two factors. First, the cubic real-gas equation of state provides increased coupling between the temperature and the partial densities compared to the perfect gas law. Second, the Soret and Dufour terms in the conservation equations represent an additional coupling between the energy and species conservation equations. Based on this evidence, the assumed PDF method is not recommended for utilization in supercritical flow simulations.

4   Conclusions

The peculiarities of supercritical mixing layers are here examined by analyzing the transitional states of four realizations differing either by the set of species, or by the perturbation wavelength used to excite the layer and achieve transition. Particularly, in contrast to gaseous mixing layers, each simulation displays concentrated regions of very high density gradient magnitude which are convoluted due to turbulence. In these regions the fluid is primarily the higher density one with small amounts of the lighter fluid dissolved into it. Departures from mixing ideality, indicating the difficulty in mixing at the molecular level (in contrast to ideal mixtures), are identified for the C_7H_{16}/N_2 layer and are found to increase with increasing density gradient. Moreover, it is shown that even in the absence of combustion, the temperature-species PDFs are well correlated (this being attributed to the higher order equation of state, and Soret and Dufour effects), meaning that their joint PDF is not properly approximated by the product of their marginal PDFs. Because under burning conditions the temperature and species will be even more correlated, this indicates that the traditional reactive flow modeling based on replacing the joint PDF representing the reaction rate by the product of the marginal PDFs, is not appropriate. This means that PDF methods applied to supercritical fluid mixing and combustion must rely on the solution of the PDF equation, as proposed by Pope [15].

References

[1] Chehroudi, B., Talley, D. and Coy, E., Initial growth rate and visual characteristics of a round jet into a sub- to supercritical environment of relevance to rocket, gas Turbine, and Diesel engines, AIAA 99-0206, 37th Aerospace Sciences Meeting, Reno, NV, 1999

[2] Chehroudi, B., Talley, D. and Coy, E., Fractal geometry and growth rate changes of cryogenic jets near the critical point, AIAA 99-2489, 35th AIAA/ASME/SAE/ASEE Joint Propulsion Conference, Los Angeles, CA, 1999

[3] Harstad, K. and Bellan, J., An all-pressure fluid drop model applied to a binary mixture: heptane in nitrogen, *Int. J. of Multiphase Flow*, 26(10), 1675-1706, 2000

[4] Keizer, J., *Statistical Thermodynamics of Nonequilibrium Processes*, Springler-Verlag, New York, 1987

[5] Mayer, W. and Tamura, H., Propellant injection in a liquid oxygen/gaseous hydrogen rocket engine, *Journal of Propulsion and Power*, 12(6), 1137-1147, 1996

[6] Miller, R. S., Harstad, K. and J. Bellan, J., Direct numerical simulations of supercritical fluid mixing layers applied to heptane nitrogen, *J. Fluid Mech.*, 436, 1-39, 2001

[7] Nomura, H., Ujiie, Y., Rath, H. J., Sato, J. and Kono, M., Experimental study on high pressure droplet evaporation using microgravity conditions, *Proc. of the Comb. Institute*, 26, 1267-1273, 1996

[8] Okong'o, N. and Bellan, J., Direct numerical simulation of a transitional supercritical mixing layer: heptane and nitrogen, submitted, 2000

[9] Okong'o, N., Bellan, J. and Harstad, K., Consistent boundary conditions for multicomponent real gas mixtures based on characteristic waves, submitted, 2000

[10] Okong'o, N., Bellan, J., Linear stability analysis of real gas two- and three-dimensional mixing layers, submitted, 2001

[11] Harstad, K., Miller, R. S. and Bellan, J., Efficient High-Pressure State Equations, *A.I.Ch.E. J.*, 43(6), 1605-1610, 1997

[12] Okong'o, N., Harstad, K. and Bellan, J.,Direct Numerical Simulations of LOX/H2 Temporal Mixing Layers under Supercritical Conditions, submitted, 2001

[13] Kim., J., Moin, P, and Moser, R. Turbulence statistics in fully developed channel flow at low Reynolds number. *J.Fluid Mech.*, vol 177, 133–166, 1987.

[14] Cortesi, A., Smith, B., Yadigarloglu, G. and Banerjee, S., Numerical Investigation of the Entrainment and Mixing Processes in Neutral and Stably-Stratified Mixing Layers. *Physics of Fluids* 11(1), 162–185, 1999

[15] Pope, S. B., PDF methods for turbulent reactive flows *Prog. Energy Combust. Sci.* 11, 119-192, 1985.

Species	m (g/mol)	T_c (K)	p_c(MPa)
Hydrogen	2.0159	33.0	1.2838
Nitrogen	28.013	126.26	3.399
Oxygen	31.9988	154.58	5.0430
Heptane	100.205	540.2	2.74

Table 1: Properties of the species investigated.

system	p_r (p)	$T_2; T_1$	ρ_2/ρ_1	$\lambda_1/\delta_{\omega,0}$	Re_0	F_{2D}	F_{3D}	Re_m
C_7H_{16}/N_2	2.17 (60 atm)	600 K; 1000 K	12.88	7.29	500	0.1	0.05	1250
C_7H_{16}/N_2	2.17 (60 atm)	600 K; 1000 K	12.88	4.57	800	0.1	0.05	1256
O_2/H_2	1.98 (100 atm)	400 K; 600 K	24.40	10.35	550	0.1	0.025	1513
O_2/H_2	1.98 (100 atm)	400 K; 600 K	24.40	7.29	750	0.1	0.025	1407

Table 2: Initial conditions of the simulations and value of the momentum thickness based Reynolds number, Re_m, at transition.

Figure 1.   Configuration of the three-dimensional mixing layer, depicted for heptane/nitrogen.

Figure 2.   One-dimensional energy spectra of the velocity fluctuations for the C_7H_{16}/N_2 simulation with $\lambda_1/\delta_{\omega,0} = 7.29$ at the transitional time ($t^* = 155$).

Figure 3.   Product thickness as a function of time for all four simulations.

Figure 4.   Conditional averages in the regions of high density gradient magnitude in the braid plane: (a) mass fractions, and (b) mass diffusion factor.

Figure 5.   Comparison of the DNS extracted PDF with the β density (using the PDF extracted moments) and the Gaussian for the C_7H_{16}/N_2 simulation with $\lambda_1/\delta_{\omega,0} = 7.29$ at the transitional time ($t^* = 155$).

Figure 6.   Comparison between the joint PDF of the partial density and temperature, and the product of the marginal PDFs for the C_7H_{16}/N_2 simulation with $\lambda_1/\delta_{\omega,0} = 7.29$ at the transitional time ($t^* = 155$).

9